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Lead-free piezoelectric low-dimension nanomaterials: controllable growth, performance optimization and exploration on micro energy harvesting devices

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Since piezoelectric effect was first noticed by Curie brothers in 1880, piezoelectric materials have attached a huge amount of attention of researchers in the worldwide. Piezoelectric effect enables the transformation from mechanical energy to electrical energy, which is able to collect the energy produced by human movements and be used to construct self-power wearable devices and pace-makers. To achieve this assumption, ZnO-based and PZT($Pb(Zr,Ti)O_3$)-based piezoelectric nanogenerators were implanted in mice and cows respectively, both of which generate voltage signals from somatic movements. However, limitations still exist since ZnO possess relatively poor piezoelectric response, while PZT contains the toxic element lead. In contrast with these two materials, KNN($(K,Na)NbO_3$)-based materials have been the most promising materials in piezoelectric field because of their good biocompatibility, excellent performance and nontoxic character.

In recent years, more and more researches were reported as energy problem has been one urging challenge to resolve. However, most of these relative studies mainly focused on bulk materials such as ceramics, while several of them were associated with low-dimension materials such as nanorods, nanowires, and nanoparticles. In our study, KNN nanorod arrays were synthesized by hydrothermal method, and the influences of reaction time on sizes, morphology, phase and piezoelectric performance of nanorods were explored. Besides, axial and radial piezoelectric responses were tested with PFM mode of atomic force microscopy, the highest radial response d_{33} is 64 pm/v. Furtherly, performance optimization was achieved after annealing in oxygen atmosphere at high temperature, which results in passivating oxygen vacancies of the surface.

However, although piezoelectric response improved a lot via annealing, it is still hard to be comparable with lead-based materials while the highest axial response d_{33} was 360pm/V after annealing at 800℃. Scholars put up an idea to improve responses by constructing MPB (morphotropic phase boundary) at room temperature. Researches pointed out that doping KNN with lithium and tantalum can decease $T_{O\text{-}T}$(orthorhombic-tetragonal temperature) to room temperature. In our study, KNN nanorods doped with lithium and tantalum were synthesized successfully on the STO($SrTiO_3$) substrates, and effects of doping content on morphology, phase and piezoelectric responses of nanorods were studied. According to the results, the radial response can be as high as 83 pm/V, which authentically improved a lot compared with pure KNN.

After systematic researches on materials synthesizing, KNN nanogenerators were assembled on the base of KNN nanorod arrays, which could output a voltage signal as high as 10V while pressed by a human finger. Furtherly, micro energy harvesting under solid conditions, energy harvesting in microfluidics and applications in soft wearable devices were accomplished.